# Bioinspired Nanomaterials: Self Stiffening Artificial Muscles


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## Abstract

Cytoskeletal organization and elasticity are greatly influenced by molecular stiffness and sterics as well as externally imposed and internally generated stresses or so it might be hypothesized [1]. These dynamic networks are generally composed of stiff filaments of actin and flexible crosslinkers [2]. Recent experiments have identified not only isotropic, nematic and raft phases of such structures but also affine and non-affine elastic regimes of protein-crosslinked actin networks. Synthetic materials lack the complexity of biological tissues, and man-made materials that respond to external stresses through a permanent increase in stiffness are uncommon. Here we report for the first time, the systems of nanotube-polydimethyl siloxane(CNT-PDMS) soft nanocomposite and analogous liquid crystalline elastomer (LCE) that mimic the actin filaments in muscle tissues. Polydomain nematic LCEs increase in stiffness by up to 90% when subjected to a low amplitude (5%), repetitive dynamic compression. Elastomer stiffening is influenced by liquid crystal content, the presence of a nematic liquid crystal phase and the use of a dynamic as opposed to static deformation. Rheological and X-ray diffraction measurements reveal that the stiffening can be attributed to a mobile nano-scale nematic director that rotates in response to dynamic compression [3]. Dynamic stiffening, not previously observed in liquid crystal elastomers may pave the way for useful development of self-healing materials and for the development of biocompatible, adaptive materials for tissue replacement.


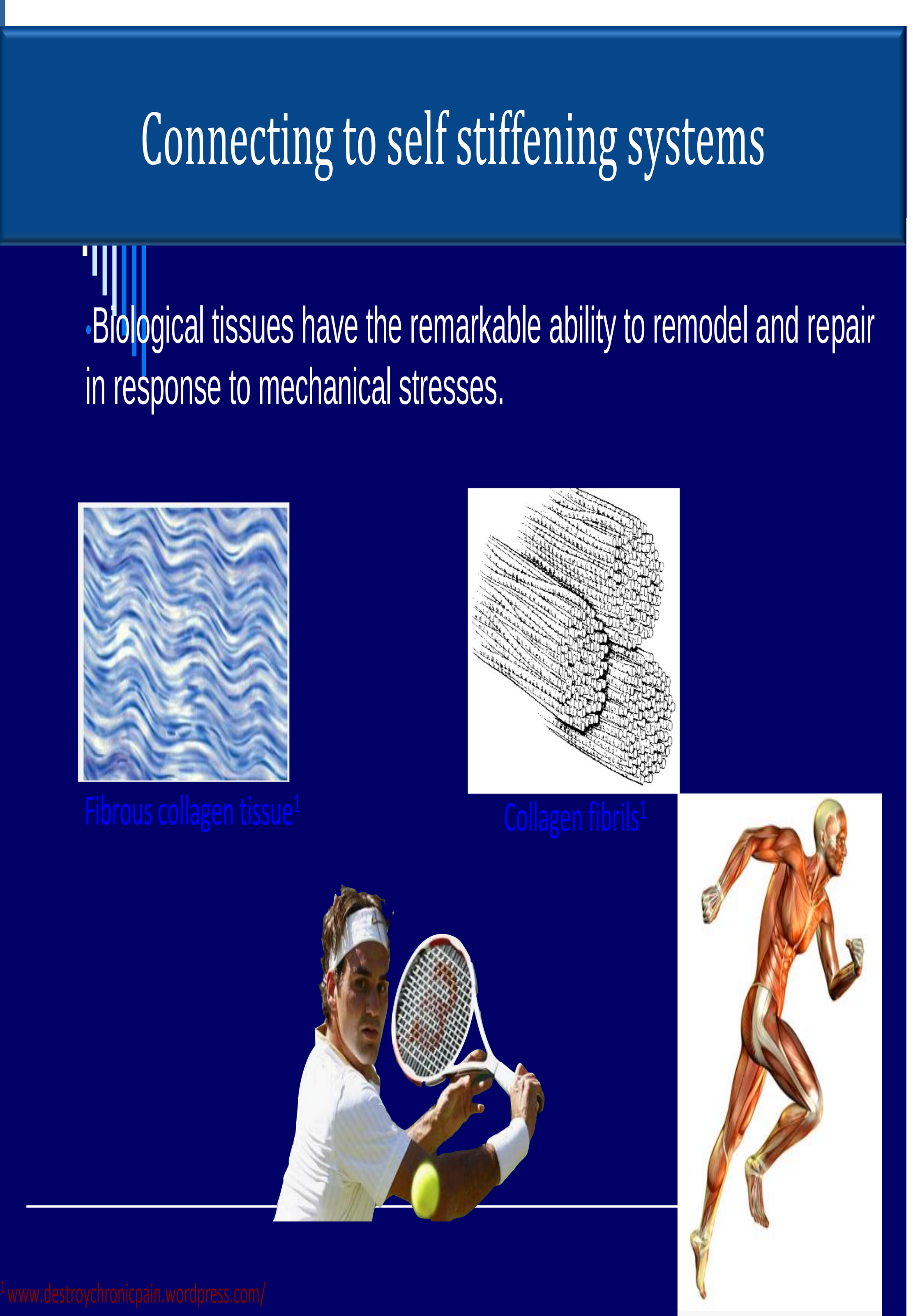


Figure 1: Self stiffened Collagen Tissues in human as adopted from the website shown in the inset.

## Experimental Observation

Most materials respond either elastically or inelastically to applied stress, while repeated loading can result in mechanical fatigue. Conversely, bones and other biomechanical tissues have the ability to strengthen when subjected to recurring elastic stress. The cyclic compressive loading of vertically aligned carbon nanotube/poly(dimethylsiloxane) nanocomposites as shown in figure 2 has revealed a self-stiffening response previously unseen in synthetic materials. This behavior results in a permanent increase in stiffness that continues until the dynamic stress is removed and resumes when it is reapplied. The effect is also specific to dynamic loads, similar to the localized self-strengthening that occurs in biological structures such as actin filaments in muscle tissues as shown in figure 5.

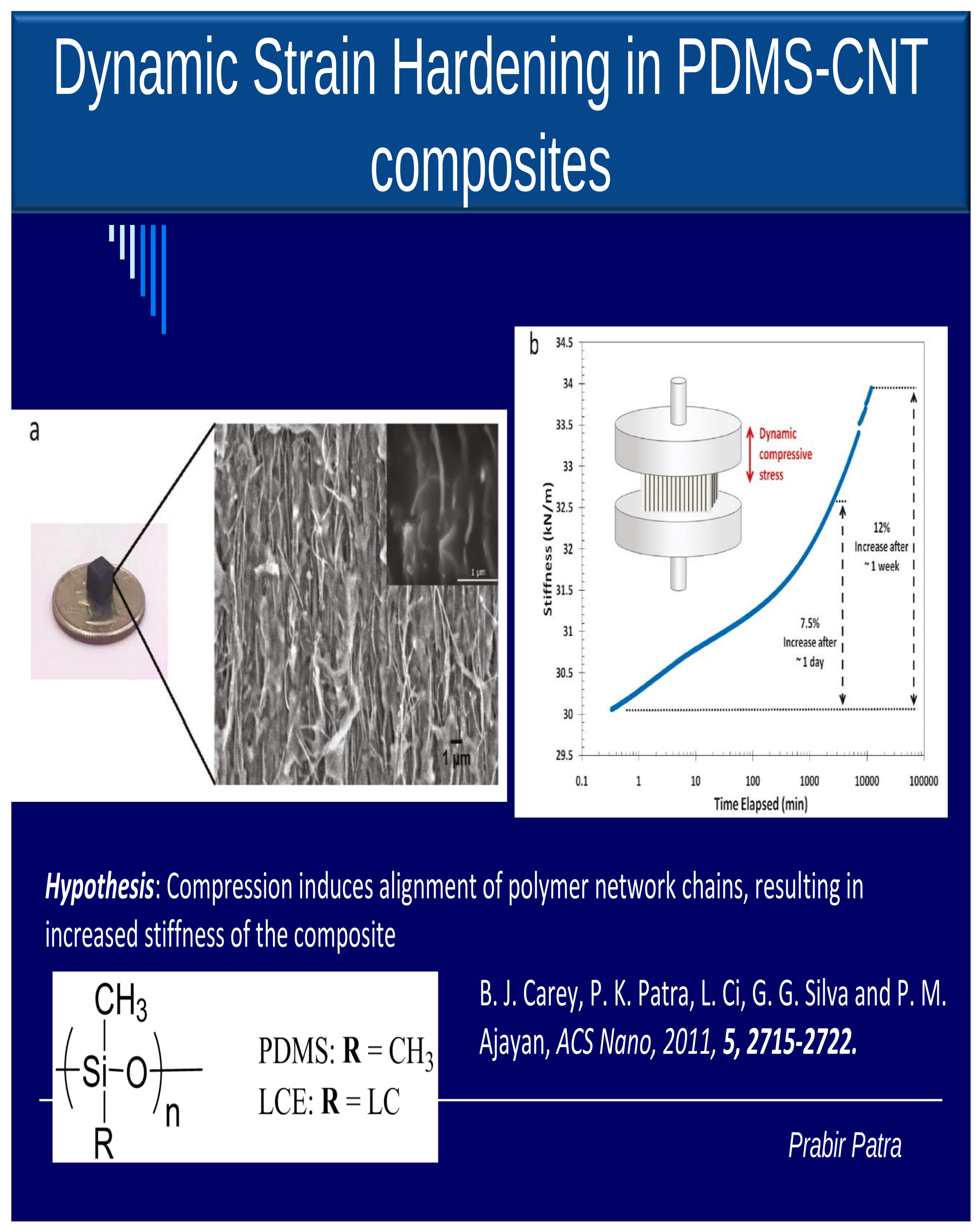


Figure 2: Compression induced alignment of PDMS chains in CNT-PDMS nanocomposite

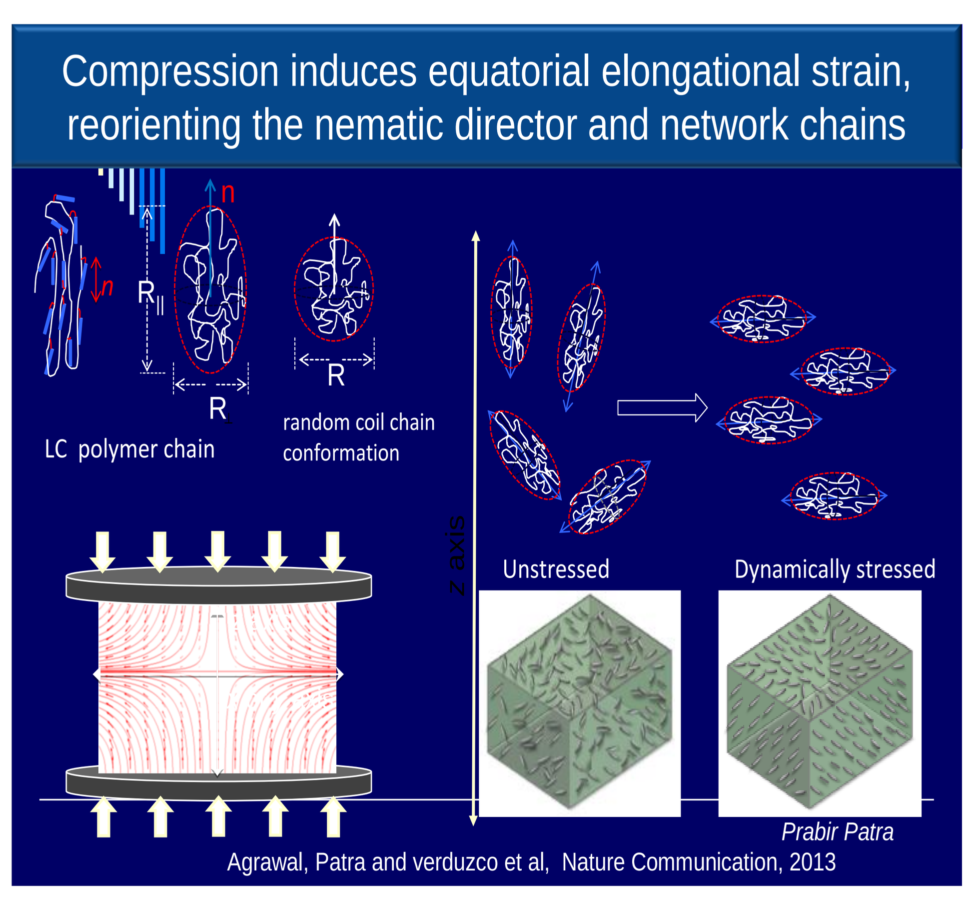


Figure 3: Compression driven reorientation of nematic director in networked LCP as experimentally evidenced in figure 6

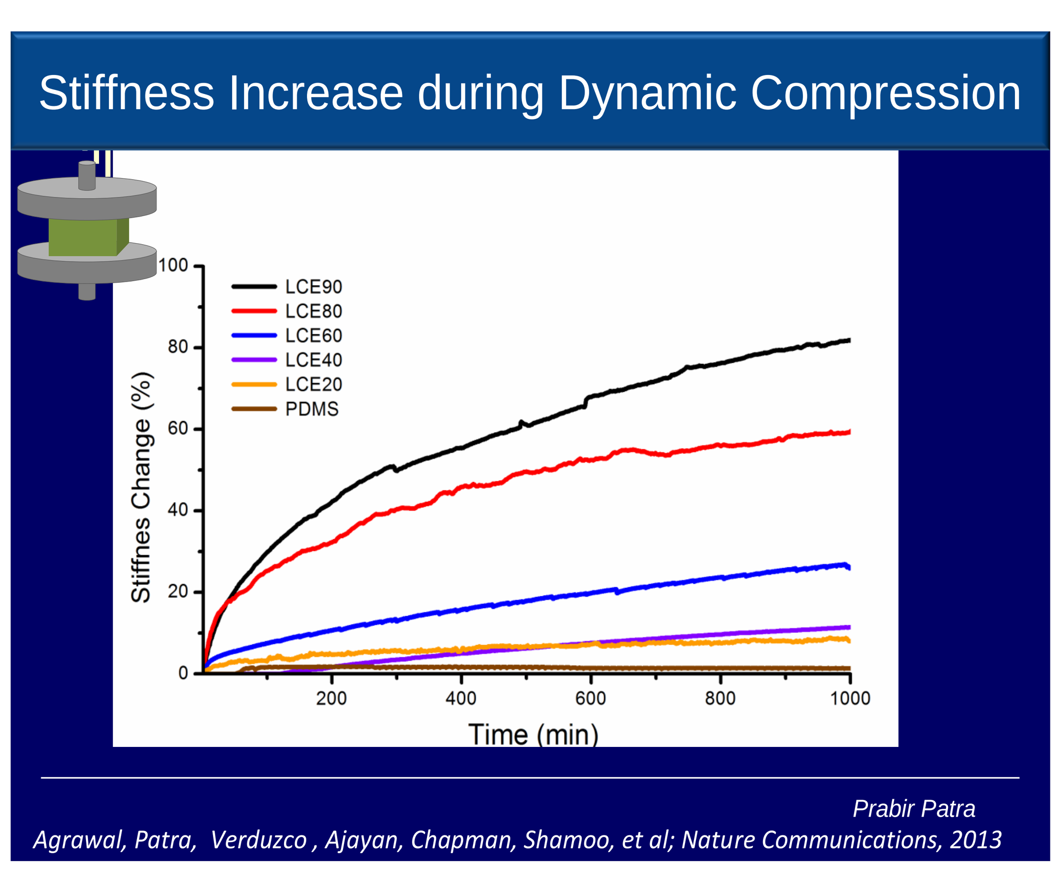


Figure 4: Dynamic compression induced increased stiffening of LCEs with varying crosslinker loading

## Discussion

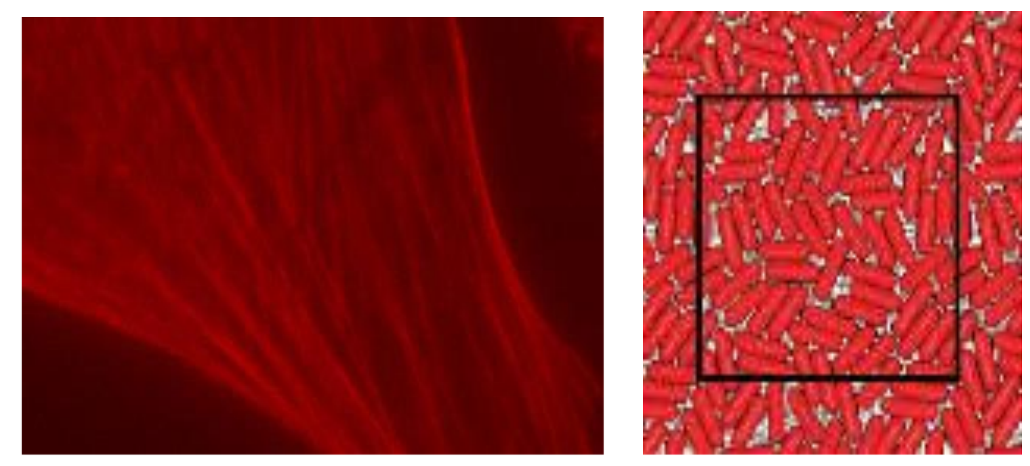

Figure 5 (left image) confocal microscopy image of actin filament (b) right image is the S parameter (cos 2ϴ =0.1) for actin filaments of LCE

Strain stiffening in LCEs contrasts with the irreversible softening of polymeric networks under cyclic strain, a phenomenon known as the Mullins effect. Although the Mullins effect is not fully understood it has been seen in crystallizable rubbers or rubbers with added fillers and has also been observed in biological tissues. Recently, stiffening behaviour was reported for bundled actin networks under cyclic shear. This was observed at higher crosslink densities and attributed to the physical nature of the network, which allowed reorganiation of the network constituents, resulting in hardening after cyclic shear. This is in contrasts with the dynamic stiffening reported here in covalent LCE networks as shown in figures 3. Figures 4 and 6 experimentally verify our observation. The novelty of the present work is the discovery of dynamic stiffening in a synthetic, homogeneous polymeric network with nano-scale liquid crystalline order. Additionally, the presence of liquid crystal order enables quantitative characterization of side-group and network chain orientation before and after deformation, establishing a direct connection between stiffening and network chain conformation.

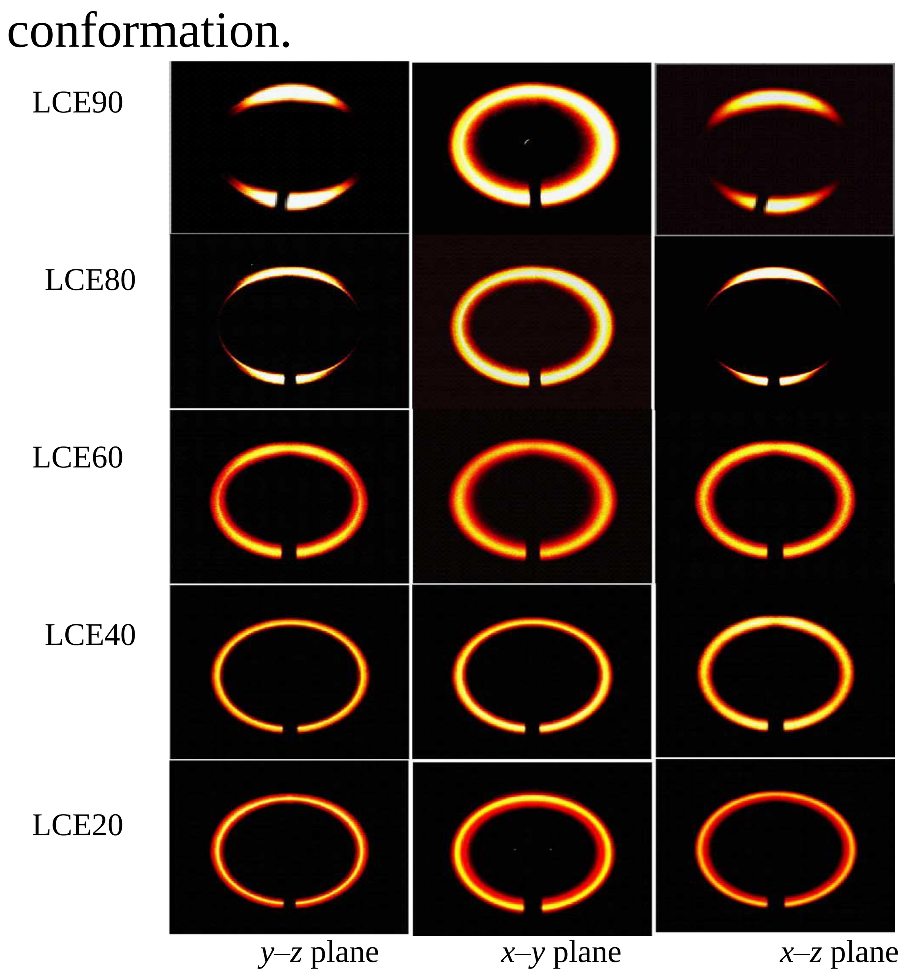


Figure 6: 2DWAXD patterns of LCEs with varying mesogenic content subjected to compressive dynamic load (5 Hz, 5% strain) for at least 16 h. The patterns are shown for three independent LCE faces in three plane directions

### Conclusions

LCE and CNT-PDMS self-stiffens in response to dynamic, compressive loading. The stiffening behaviour observed here is for a permanent network at low strains and can be attributed to a mobile nematic director. Director reorientation and alignment at low strains and dynamic compression has not been previously reported in LCEs and suggest underlying network relaxation modes at 5 Hz, which governs the response. LCEs that increase in stiffness may be useful for the development of self healing materials and for the development of biocompatible, adaptive materials for tissue replacement which requires orinetation driven actin filament formation.


## References:

1. Speicher, D.W. & Marchesi, V. T. Erythrocyte spectrin is comprised of many homologous triple helical segments. *Nature* **311**, 177–180 (1984).
2. Gardel, M. L. *et al*. Elastic behavior of cross-linked and bundled actin networks. *Science* **304**, 1301–1305 (2004).
3. Agarwal A. *et al.* Dynamic self stiffening in liquid crystalline elastomer, *Nature Communications* , **4**, 1739 (2013)